WEST Search History

DATE: Sunday, June 15, 2003

Set Name		Hit Count	Set Name
side by side			result set
DB=DWPI; $PLUR=YES$; $OP=OR$			
L15	(phenol) and (methacrylic adj acid) and (separator or (fuel adj cell))	4	L15
L14	(bis adj phenol) and (methacrylic adj acid) and (separator or (fuel adj cell))	0	L14
L13	(bisphenol) and (methacrylic adj acid) and (separator or (fuel adj cell))	0	L13
DB=JPAB; $PLUR=YES$; $OP=OR$			
L12	((bisphenol or toto) and (methacrylic adj acid)) and (separator or (fuel adj cell))	0	L12
DB=EPAB; $PLUR=YES$; $OP=OR$			
L11	((bisphenol or toto) and (methacrylic adj acid)) and (separator or (fuel adj cell))	0	L11
DB=DWPI; PLUR=YES; OP=OR			
L10	((bisphenol or toto) and (methacrylic adj acid)) and (separator or (fuel adj cell))	0	L10
DB=USPT; PLUR=YES; OP=OR			
L9	((bisphenol or toto) and (methacrylic adj acid)) and (separator or (fuel adj cell))	414	L9
L8	L7 and (separator or (fuel adj cell))	8	L8
L7	L4 and ((bisphenol or toto) and (methacrylic adj acid)).clm.	353	L7
L6	L5 and (separator or (fuel adj cell))	. 8	L6
L5	L4 and ((bisphenol or toto) and methacrylic).clm.	399	L5
L4	L3 and styrene	7330	. L4
L3	L2 and epoxy	8812	L3
L2	(bisphenol or toto) and methacrylic	12877	L2
L1	(biphnol or toto) and methacrylic	213	L1

END OF SEARCH HISTORY

WEST

End of Result Set

Generate Collection Print

L1: Entry 3 of 3

File: USPT

Jun 26, 2001

DOCUMENT-IDENTIFIER: US 6251308 B1

** See image for Certificate of Correction **

TITLE: Highly conductive molding compounds and $\underline{\text{fuel cell}}$ bipolar plates comprising these compounds

Abstract Text (1):

A conductive polymer is disclosed which is suitable for use in applications which require corrosion resistance including resistance to corrosion when subjected to acidic flow at temperature ranging from -40 to 140 degrees Fahrenheit and which can be molded into highly intricate and thin specimens which exhibit consistent conductivity, sufficient strength and flexibility, and appropriate surface characteristics. In particular the invention involves molding unsaturated prepolymer resin composition which have high loadings of conductive fillers. Further to enable the necessary characteristics, the compositions include rheological modifiers such as Group II oxides and hydroxides; carbodiamides; aziridines; polyisocyanates; polytetrafluoroethylene (PTFE); perfluoropolyether (PFPE), and polyethylene. Ostensibly, these modifiers act to alter the apparent molecular weight and three dimensional prepolymer network structures correcting rheological deficiencies which otherwise lead to excessive resin particulate separation during the molding process and large variances in bulk conductivity across the plate surface. The composition is disclosed for use in electrochemical cells, such as fuel cells.

Brief Summary Text (5):

One area in particular where it would be beneficial to solve the previously mentioned strength, durability, and molding issues is for application in <u>fuel cells</u>. Electrochemical <u>fuel cells</u> have great appeal as a potentially limitless energy source that is clean and environmentally friendly. These <u>fuel cells</u> can, in addition, be constructed at an appropriate scale for small scale energy consumption, such as household use, or for industrial scale use, and even for commercial power generation: They have portable applications to power small appliances (such as computers or camping equipment), or automobiles and other forms of transportation. Although these different applications involve differences in size, the fundamental construction remains the same for generation of power varying from less than one to a few thousand kilowatts.

Brief Summary Text (6):

Basically, a <u>fuel cell</u> is a galvanic cell in which the chemical energy of a fuel is converted directly into electrical energy by means of an electrochemical process. The fundamental components of the <u>fuel cell</u> are an electrode comprising an anode and a cathode, eletrocatalysts, and an electrolyte. Work has been done in perfecting both liquid and solid electrolyte <u>fuel cells</u> and the present invention may find use in both types of fuel cells.

Brief Summary Text (7):

Solid electrolytes include polymeric membranes which act as proton exchange membranes typically fueled by hydrogen. These membranes usually comprise a perfluorinated sulphonic acid polymer membrane sandwiched between two catalyzed electrodes that may utilize platinum supported on carbon as an electrocatalyst. Hydrogen <u>fuel cells</u> form a reaction chamber, which consumes hydrogen at the anode. At the cathode, oxygen reacts with protons and electrons at the electrocatalytic sites yielding water as the reaction product. A three-phase interface is formed in

the region of the electrode and a delicate balance must be maintained between the electrode, the electrolyte, and the gaseous phases.

Brief Summary Text (8):

Systems involving the use of other electrolytes have been also been studied. These would include alkaline $\underline{\text{fuel cells}}$, phosphoric acid $\underline{\text{fuel cell}}$, molten carbonate $\underline{\text{fuel cells}}$, and solid oxide $\underline{\text{fuel cells}}$. However, the principles are similar, as are some of the issues in perfecting these products.

Brief Summary Text (9):

A fuel cell reactor may comprise a single-cell or a multi-cell stack. In any case, the cell includes at least two highly conductive flow field plates that serve multiple functions. These plates may function as current collectors that provide electrical continuity between the <u>fuel cell</u> voltage terminals and electrodes. They also provide mechanical support (for example for the membrane/electrode assembly). In addition, these plates act to transport reactants to the electrodes and are essential to establishing the previously mentioned delicate phase balance.

Brief Summary Text (10):

Typically, the fuel cell plates are thin relatively flat plate members that include a highly complex network of interconnecting channels that form the flow field area of the plate. The configuration of these channels is highly developed in order to maintain the proper flow of reactants and to avoid channeling or the formation of stagnant areas, which results in poor <u>fuel cell</u> performance. It is critical that the flow of the reactants is properly managed, and that the electrocatalysts are continuously supplied with precisely the appropriate balance of reactants. Thus, it is essential for the plates to define and maintain clear passages within the highly engineered flow labyrinth. Moreover, in order to assure a satisfactory life, the plates must be able to resist surface corrosion under a variety of conditions. For example, fuel cells may be placed outside and subject to ambient weather. Thus, the cells must be resistant to stress cracking and corrosion at temperature ranging from -40 to 200 degrees Fahrenheit. Further, since the conditions within the cell are corrosive, the cells must also be resistant to chemical attack at these temperatures from various corrosive substances. For example, the plates may be subjected to de-ionized water, methanol, formic acid. formaldehyde, heavy naptha, hydrofluoric acid, tertafluoroethylene, and hexafluoropropylene depending on the fuel cell type. Moreover, the conditions within the <u>fuel cell</u> may lead to elevated temperatures, i.e. from 150 to 200 degrees Fahrenheit, as well as elevated pressures, i.e. from ambient to 30 p.s.i. Corrosive decomposition needs to be avoided since it almost certainly would cause a system failure by changing the flow patterns within the fuel

Brief Summary Text (11):

Past attempts at solving the various requirements for <u>fuel cell</u> plates have included the use of metal and machined graphite plates. The use of metal plates result in higher weight per cell, higher machining costs and possibly corrosion problems. Machined graphite plates solve the weight and corrosion problems but involve high machining cost and result in fragile products, especially when prepared as very thin plates. Some use of graphite/poly(vinylidene fluoride) plates has been made but these have been characterized as being expensive and brittle and having long cycle times.

Brief Summary Text (12):

U.S. Pat. No. 4,197,178 is incorporated herein for its teaching of the working and compositions of electrochemical cells. U.S. Pat. No. 4,301,222 is incorporated herein for its teachings on graphite based separators for electrochemical cells.

Brief Summary Text (16):

The foregoing improvements in specimens molded from these compositions enable the low cost mass production of bipolar plates as an additional embodiment of the invention. These could be used for portable <u>fuel cells</u>, as well as stationary power units.

Drawing Description Text (2):

FIG. 1 is an illustration of a fuel cell assembly utilizing a bipolar cell plate;

and

Drawing Description Text (3):

FIG. 2 is an illustration of a bipolar <u>fuel cell</u> plate that can be made in accordance with the present invention.

Detailed Description Text (5):

In general, the vinyl ester resins that can be used are the reaction products of epoxy resins and a monofunctional ethlenically unsaturated carboxylic acid. More specifically, these vinyl ester resins are the reaction product of an epoxy terminated oligomer, for example, an epoxy functionalized <u>bisphenol</u> A with an acrylic acid, or <u>methacrylic</u> acid forming acrylic terminal groups on the oligomer. The vinyl esters have predominantly terminal unsaturation while the unsaturated polyesters have predominantly internal unsaturation.

Detailed Description Text (7):

Another component to the molding composition is fillers. In accordance with the invention the predominant filler is a conductive filler in order to impart electrical conductivity of the final molded product. A preferred filler is graphite particles, in particular, a synthetic crystalline graphite particle, such as currently supplied by Asbury Graphite in Asbury, N.J. under the designation Asbury 4012. This graphite is characterized as having less than 10% particles greater than 150 microns and less than 10% smaller than 44 microns in diameter. Other graphite fillers include: Ashbury A99, Ashbury 3243, Ashbury modified 4012, Ashbury 3285, Ashbury 230U; TimrexR KS 75 and 150, and TimrexR KC 44, all sold by TIMCAL of Westlake, Ohio; and Calgraph Sold by SGL Technic Inc of Valencia, Calif. This filler is used at a loading of at least 50% by weight. Other conductive fillers such as other forms of graphite (including graphite pitch-based fibers), metal particles, or metal coat particles may be used in conjunction with the graphite filler, or even alone. Desirably conductive fillers are at least about 50, about 60, or about 65 weight percent of the molding composition. More desirably the filler is more than about 70 or 71 percent to about 78 weight percent of the molding composition. Alternatively this amount can be expressed as at least about 250 phr, more preferably at least about 275, or even 300 phr. Alternatively stated the conductive fillers are present in an effective amount to result in a bulk conductivity of at least about 40, about 50, about 60, about 70, about 80, about 85, about 90 or about 96 S/cm when measured in accordance with ASTM Test Standard No. F1529-97 for a molded article having a thickness from about 0.060 to about 0.200 inches. Current technology in fuel cell plates uses a bulk conductivity of at least about 55, and preferably at least about 70.

Detailed Description Text (9):

An essential component to the improved molding composition is a rheological modifier, which may act to increase the molecular weight such as by chain extension of the resin prepolymer. Suitable modifiers include Group II oxides and hydroxides, such as calcium or magnesium oxide; carbodiamides; aziridines; and polyisocyanates. It is believed that the foregoing modifiers act chemically by co-reacting into the polymer backbone at carboxy or hydroxy sites. Other suitable modifiers include therefore polytetrafluoroethylene (PTFE); perfluoropolyether (PFPE), and polyethylene. These modifiers may act to reduce shear and thus promote flow in the composition during molding. Fumed silica is an example of a substance which may act mechanically to increase molding viscosity and therefore be a suitable rheological modifier for this invention. Combinations of two or more rheological modifiers may be desirable for optimum properties. In this application they are used to modify the resin structure to prevent phase separation of the resin from the conductive filler (in particular in view of the high loadings of the conductive filler, i.e. over 50% or even 65% by weight or more of graphite) The modifiers are further used in general to enable the achievement of a high definition conductive polymeric fuel cell plate.

<u>Detailed Description Text</u> (13):

Molded products made from the compositions of the present invention are useful for a variety of applications demanding complex configurations, conductivity, as well as strength, and corrosion resistance. One particularly advantageous product which can be made by compression molding is a bipolar plate for use in <u>fuel cells</u>. An example

of such a plate is shown in FIG. 1. The drawing of this plate is intended to illustrate the molding capabilities of the conductive compound of the present invention. It is not necessarily intended to provide optimal, or even operative, field flow design. It should not limit the invention in any way. The plate 10 includes a fluid flow face with one or more generally parallel and or serpentine flow channels 12. The flow channels receive and transmit fluids through ports 14 and 16 which are in fluid communication with corresponding entry and exit fluid manifolds 18 and 19. The plate has a dimension which will vary from 1 to 20 inches in length and width, and having a thickness of 0.02 to 0.3 inch, with a cross-sectional depth of the flow channel in the range of about 0.005 to 0.080 inch. The cross-sectional width of a land separating adjacent flow channel sections is in the range of 0.01 to 0.1 inch. The plate may include a number of peripheral through

Detailed Description Text (14):

holes 20 that act as a manifold for fuel transportation.

FIG. 2 illustrates the unassembled components of a fuel cell. This fuel cell has a --base unit 12 which includes debossed means to accept a reformer 14 and a fuel cell stack 16 which is comprised of a plurality of bipolar plates 20 which are sandwiched between a stack cap 22 and a stack base 24. The fuel cell further includes a heat exchanger 26. An enclosure 30 provides a leak-proof housing for the unit.

Detailed Description Text (18):

Resin B is Atlac 382ES from Reichhold Chemicals, Inc. in Research Triangle Park, N.C. It is characterized as a bisphenol fumarate resin. It was diluted to about 55 wt. % solids with styrene.

Detailed Description Text (19):

Resin C is Dion 6694 diluted to 55 wt. % solids in styrene. It is available from Reichhold Chemicals, Inc. It is characterized as a modified bisphenol fumarate polyester.

Detailed Description Text (23):

Resin G is 9100 from Reichhold Chemicals, Inc. It is characterized as a bisphenol-epoxy vinyl ester. It was diluted to 54-58 wt % solids with styrene.

Detailed Description Text (54):

The molding parameters for the molding compositions are as follows: Molding temperature for plaques was 295.degree. F. with a molding time of 3 minutes and a charge weight of 173 g. The molding temperature for prototype bipolar plates was 290.degree. F. with a molding time of 3 minutes and a charge weight of 300 g. It was observed that the use of specific thermosetting resins with a conductive filler in combination with various rheological additives (thickeners) improved the bipolar plate composition in regards to having a product which can be used in mass production of electrochemical, e.g. fuel, cell bipolar plates.

CLAIMS:

- 10. A composition as set forth in claim 4 wherein said conductive filler is a synthetic crystalline graphite particle and said unsaturated prepolymer resin is one or more resins selected from the group consisting of epoxy vinyl resin, bisphenol fumarate resin, modified bisphenol fumarate polyester resin, unsaturated polyester resin, urethane-modified vinyl ester resin, bisphenol-epoxy vinylester resin, elastomer-modified vinyl ester resin, epoxy novolac vinyl ester resin and unsaturated isocyanurate vinyl ester resin.
- 26. An electrochemical cell flow field plate as set forth in claim 20 wherein said conductive filler is a synthetic crystalline graphite particle and said unsaturated prepolymer resin is one or more resins selected from the group consisting of epoxy vinyl resin, bisphenol fumarate resin, modified bisphenol fumarate polyester resin, unsaturated polyester resin, urethane modified vinyl ester resin, urethane-modified vinyl ester resin, <u>bisphenol</u>-epoxy vinylester resin, elastomer-modified vinyl ester resin, epoxy novolac vinyl ester resin and unsaturated isocyanurate vinyl ester
- 34. A conductive molding composition as set forth in claim 33 wherein said

prepolymer resin is selected from the group consisting of epoxy vinyl resin, bisphenol fumarate resin, modified bisphenol fumarate polyester resin, unsaturated polyester resin, urethane modified vinyl ester resin, urethane-modified vinyl ester resin, bisphenol-epoxy vinylester resin, elastomer-modified vinyl ester resin, epoxy novolac vinyl ester resin and unsaturated isocyanurate vinyl ester resin.

49. A molded conductive product as set forth in claim 45 wherein said unsaturated prepolymer resin is one or more resins selected from the group consisting of epoxy vinyl resin, <u>bisphenol</u> fumarate resin, modified <u>bisphenol</u> fumarate polyester resin, unsaturated polyester resin, urethane modified vinyl ester resin, urethane-modified vinyl ester resin, <u>bisphenol</u>-epoxy vinylester resin, elastomer-modified vinyl ester resin, epoxy novolac vinyl ester resin and unsaturated isocyanurate vinyl ester resin.